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Rapid Communications

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Surface phonon dispersion on Al(100)

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The first experimental measurement of the Rayleigh mode dispersion curve for the Al(100) surface is reported. Excellent agreement is found between the experimental results and the theoretical first-principles calculation performed by Bohnen and Ho [J. Vac. Sci. Technol. A **5**, 462 (1987)] for the phonon energy at $\bar{X}$. This is to be contrasted with the large discrepancy reported by Toennies and Wöll [Phys. Rev. B **36**, 4475 (1987)] between the experimental and theoretical values at $\bar{X}$ for the Al(110) surface.

Experimental and theoretical surface phonon studies of clean metal surfaces have intensified in the past few years. Among these studies, by far the most interesting and controversial were those that were concerned with the (111) surfaces of the noble metals Cu, Ag, and Au, which were obtained by high-resolution He-atom time-of-flight spectroscopy.[1] These He-atom scattering measurements determined the dispersion of an anomalous longitudinal surface phonon resonance and the Rayleigh mode. Bortolani, Franchini, Nizzoli, and Santoro,[2] who used lattice dynamical models, interpreted these He-atom scattering results in terms of a 50% softening of the intralayer surface force constants. Jayanthi, Bliz, Kress, and Benedek[3] have also considered the above experimental results and argued that both the anomalous surface resonance and the softening of the surface force constants are related to a surface charge redistribution that increases the Coulomb attraction at the surface, thereby effectively reducing the ion-ion repulsion there.

The study of a free-electron-like metal, such as aluminum, should provide insight into the discussion surrounding the role of the *sp-d* hybridization in the noble metals.[4] Furthermore, the surface phonon energies derived from first-principles calculations at $\bar{X}$ for Al(100) and at $\bar{X}$ and $\bar{Y}$ for Al(110) have been tabulated.[5] The calculations are thought to be reliable because they correctly reproduce both the multilayer relaxation of the Al(110) surface and the absence of any significant relaxation of the Al(100) surface. At the time the above calculations were made, there were no experimental results for comparison. Recently, Toennies and Wöll[4] have reported the surface phonon dispersion curves for Al(110) in the $\bar{\Gamma}$-$\bar{X}$ and $\bar{\Gamma}$-$\bar{Y}$ directions of the surface Brillouin zone (SBZ). A comparison of the experimental and theoretical values at $\bar{X}$ showed a discrepancy of approximately 2.5 meV. Because of the extent of the discrepancy, Toennies and Wöll[4] suggested that there might be a systematic error in the theory. On the other hand, as was mentioned earlier, the theory is in excellent agreement with the previous low-energy electron-diffraction (LEED) study of the multilayer relaxation of Al(110).[6]

The purpose of this Rapid Communication is to present the surface phonon dispersion curves for Al(100) in the $\bar{\Gamma}$-$\bar{X}$ direction of the SBZ in order to further test the theoretical calculation. We believe the Al(100) surface provides a more reliable test for the theory since it is more amenable to surface preparation techniques than the Al(110) surface which is known to contain small disordered domains which are misaligned with respect to each other.[6]

The measurements were performed in a diffusion-titanium sublimation pumped ultrahigh-vacuum system (base pressure $\simeq 5\times10^{-11}$ Torr). The Al(100) sample was cleaned by cycles of Ar^+ ion bombardment and annealing at 700 K. Auger analysis shows the presence of trace amounts of carbon and oxygen, but the LEED pattern is of high quality. The ratio of the Auger signals for oxygen (510 eV) and Al (68 eV) is less than 0.001. Others[7] reported a similar ratio for nominally clean aluminum surfaces. The high-resolution electron-energy-loss spectra (HREELS) taken at an incident energy of 8.8 eV show only a small broad peak at $\simeq 810$ cm^{-1}, indicating the presence of subsurface oxygen contamination.[8] However, the intensity of this oxygen loss peak relative to the elastic peak is less than 0.1% and is comparable to the corresponding relative intensities reported previously for nominally clean Al(110).[8,9] The HREELS measurements were obtained with the spectrometer described pre-

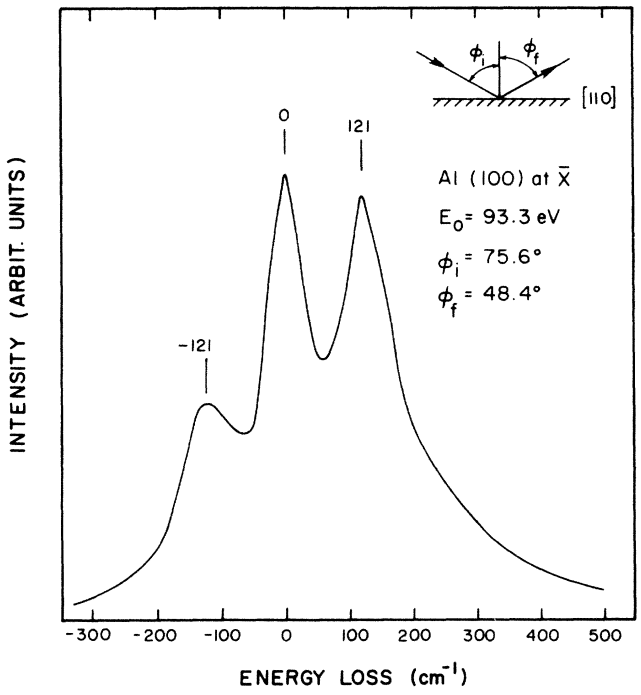

FIG. 1. High-resolution electron-energy-loss spectrum showing the Rayleigh mode.

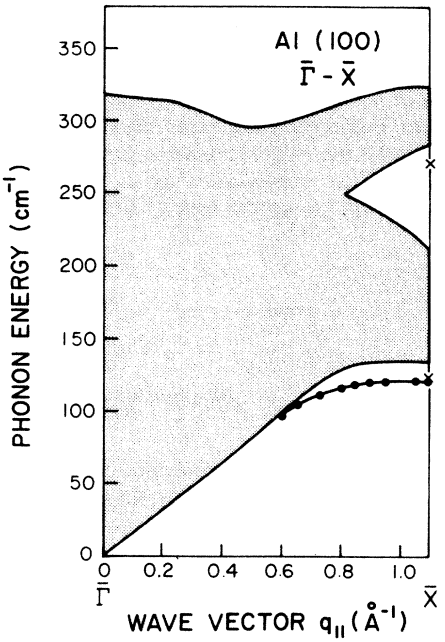

FIG. 2. Phonon dispersion curves in the $\hat{\Gamma}$-$\hat{X}$ direction of Al(100). Experimental points for the Rayleigh mode are denoted by filled circles; crosses indicate the results of the calculation (Ref. 5).

viously[10] having a nominal energy resolution of 5 meV for incident energies in the range of 1–240 eV. The scattering plane was defined by the [001] and [110] directions.

Figure 1 presents the HREELS spectra at the $\bar{X}$ point of the SBZ ($q_{\parallel}=1.097$ Å^{-1}). In addition to the peak at 0 cm^{-1}, which corresponds to the diffuse elastic electron scattering, there are two other peaks at ± 122 cm^{-1} corresponding to the phonon loss and gain, respectively, and assigned to the Rayleigh mode (S_4). Impact scattering selection rules[11] preclude the observation of the S_1 mode for this scattering plane. Figure 2 presents the dispersion of the Rayleigh mode, peak energies being determined by a least-squares fit to Gaussians based upon a series of measurements. The energy of the Rayleigh mode at $\bar{X}$ is determined to be 122 ± 5 cm^{-1}, which is in excellent agreement with the value of 123 cm^{-1} predicted for this surface.

In summary, we have presented the first experimental results for the dispersion of the Rayleigh mode for the Al(100) surface. Agreement between experimental and theoretical values of the surface phonon energy at $\bar{X}$ is excellent. This suggests that the first-principles calculation of surface phonon energies for other metals could be very valuable.

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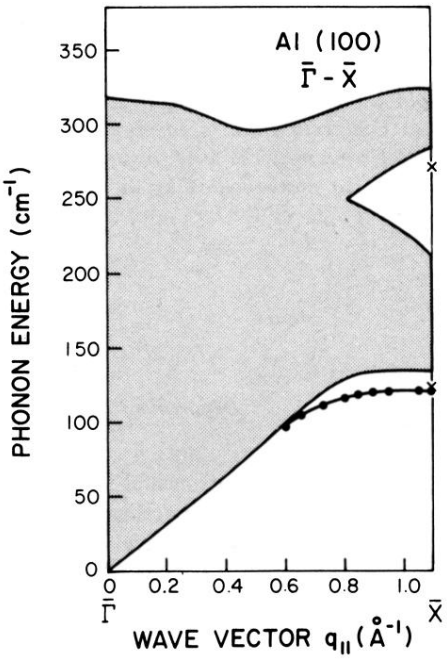

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